

# Spatial and temporal variations of N<sub>2</sub>O emissions from global forest and grassland ecosystems

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## ABSTRACT

A process-based dynamic ecosystem model of TRIPLEX-GHG was used to estimate the spatial and temporal patterns of N<sub>2</sub>O fluxes from global forest and grassland ecosystems under the effects of global warming and elevated CO<sub>2</sub> concentrations. From 1992 to 2015, the estimated average N<sub>2</sub>O emissions from forests and grasslands were  $3.62 \pm 0.16$  Tg N yr<sup>-1</sup> and  $1.40 \pm 0.03$  Tg N yr<sup>-1</sup>, respectively. Tropical regions made large contributions (83.9% for forests and 74% for grasslands) to the total N<sub>2</sub>O budgets, which were due to the larger N<sub>2</sub>O flux values and large natural forest and grassland areas. The regional variations in N<sub>2</sub>O emissions mainly resulted from the differences in the spatial distributions of climate characteristics, especially the precipitation patterns. In addition, anomalous years when N<sub>2</sub>O emissions were relatively low/high were mainly due to the changes in climate patterns, which may have been induced by El Niño/La Niña events with different strengths and frequencies. Soil N<sub>2</sub>O emissions from forests showed a positive effect on the atmospheric N<sub>2</sub>O concentrations during June to November ( $R^2$ : 0.14–0.28), while those from grasslands showed a positive effect during the growing seasons ( $R^2$ : 0.17–0.28). Although natural N<sub>2</sub>O sources (forests and grasslands in this study) showed slightly increasing trends, with 9.9 Gg N increment per year for forests and 2.1 Gg N increment per year for grasslands, they were not the main contributors to the elevated N<sub>2</sub>O concentrations.

## 1. Introduction

Nitrous oxide (N<sub>2</sub>O) is a principal greenhouse gas (GHG) that has a relative global warming potential that is 298 times that of CO<sub>2</sub> over a 100-yr period (IPCC, 2013). Moreover, N<sub>2</sub>O contributes approximately 7% to radiative forcing (Artaxo et al., 2007), and it is also one of the largest ozone-depleting substances emitted from the biosphere (Ravishankara et al., 2009). Atmospheric N<sub>2</sub>O has increased by 21% compared to the preindustrial level, and N<sub>2</sub>O concentration reached 325.9 ppb per year in 2013 (Ciais et al., 2014). Natural N<sub>2</sub>O emissions play important roles in determining the total emissions and feedback between the atmosphere and biosphere. N<sub>2</sub>O emissions from natural forests and grasslands account for approximately 15–55% and 9–20% of the total N<sub>2</sub>O emissions, respectively (Tian et al., 2013, 2016; Xu and Prentice, 2008; Zhuang et al., 2012). Furthermore, natural N<sub>2</sub>O emissions are closely connected with climatic and ecological variables. The

complex interactions among emissions, climate and ecological variables has contributed to the large uncertainty in the estimations of N<sub>2</sub>O emissions from natural soils (Butterbach-Bahl et al., 2013; Chatskikh et al., 2005). Additionally, climatic and ecological variables also constitute potentially important feedbacks in the global earth system; for example, terrestrial N<sub>2</sub>O emissions are enhanced under warmer climates and higher atmospheric CO<sub>2</sub> concentrations (van Groenigen et al., 2011; Xu et al., 2012). The associated feedback loop amplifies anthropogenic climate change and is reflected in paleontological records on glacial–interglacial and centennial timescales (Cai et al., 2014; Stocker et al., 2013). These potential roles of natural N<sub>2</sub>O emissions in the climate system highlight the importance of the scientific understanding of the key processes that govern the production of emissions as well as accurate predictions of changes in emissions resulting from a changing climate.

Plenty of in situ N<sub>2</sub>O experiments have been conducted throughout

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the world over the past several decades (Williams et al., 1992; Abalos et al., 2016; Zhuang et al., 2012; Zhang et al., 2017); however, up-scaling field observations from a site level to a regional or global scale is a great challenge. Several upscaling studies on N<sub>2</sub>O emissions have been published using different approaches, such as the flux empirical extrapolation method (Zhuang et al., 2012), inverse method (Thompson et al., 2013) and process-based models (Zhang et al., 2017; Tian et al., 2018). Neither extrapolation nor inverse methods provide insight about the processes that are primarily responsible for GHG emissions or how to adequately characterize the full heterogeneity present in the landscape; therefore these methods involve large uncertainties when estimating regional emissions. In contrast, process-oriented models are often based on a better understanding of the biogeochemistry of GHG production and consumption (e.g., nitrification and denitrification); these models are considered powerful research tools for estimating regional and global N<sub>2</sub>O emissions (Tian et al., 2018). Some global N<sub>2</sub>O models attempt to simulate the global emissions of N<sub>2</sub>O by considering a variety of complex regulating parameters or synthesizing the available flux measurements and known sources (Kiese et al., 2003, 2005; Li et al., 1992; Potter et al., 1996; Saikawa et al., 2013; Tian et al., 2013; Werner et al., 2007; Xu et al., 2012). However, there are still large uncertainties in terms of model calibration and validation, and large divergences exist when simulating the spatial and temporal variations of N<sub>2</sub>O emissions with different models (Tian et al., 2013).

A better understanding of the N<sub>2</sub>O emissions from natural forests and grasslands and how these emissions may change over time could help us better determine the impacts of N<sub>2</sub>O and help policy-makers make better decisions when they are debating regulations for anthropogenic sources of N<sub>2</sub>O. TRIPLEX-GHG model has already been calibrated and validated in simulating N<sub>2</sub>O emissions across different ecosystems and latitudes in our previous study (Zhang et al., 2017). This study is the first application of this model for global N<sub>2</sub>O estimation. The specific objectives of this study include 1) providing an updated estimate of N<sub>2</sub>O budgets for global natural forests and grasslands using the TRIPLEX-GHG model, which may help narrow down the ranges of N<sub>2</sub>O emission estimations from terrestrial ecosystems; 2) simulating temporal and spatial patterns of N<sub>2</sub>O emissions under the impacts of multiple climate factors and analyzing the primary cause of N<sub>2</sub>O emission anomalies; 3) exploring the relationships between soil N<sub>2</sub>O emissions from natural forests and grasslands and the atmospheric N<sub>2</sub>O concentrations.

## 2. Methodologies and model

### 2.1. Data description

In this study, we applied a series of spatiotemporal data sets to represent environmental changes at a spatial resolution of  $0.5^\circ \times 0.5^\circ$  latitude/longitude from 1992 to 2015. These data include daily climate conditions, such as the minimum, average and maximum temperature, precipitation, specific humidity, air pressure and wind speed. The climate data were downloaded from the CRUNCEP website (<https://www.earthsystemgrid.org/dataset/ucar.cgd.cesm4.CRUNCEP.v4.TPHWL6Hrly.html>). The soil classification map used in this study included soil texture (clay, sand and silt fraction) and soil pH and was based on the Digital Soil Map of the World (DSMW), which we obtained from the FAO/UNESCO Soil Map of the World (<http://www.fao.org/soils-portal/soil-survey/soil-maps-and-databases/faunesco-soil-map-of-the-world/en/>) and conjoined the DSMW attributes with the soil properties dataset using methods in Batjes (2006). The soil C data and soil C:N ratio data were adopted from the global soil dataset (IGBP-DIS; 2000). The topographic input data were generated based on a global digital elevation model (DEM) with an approximate spatial resolution of 1 km (GTOPO30). The annual vegetation maps were aggregated from the annual maps from the Climate Change Initiative land cover project led by the European Space Agency (ESA – CCI – LC), which span cover a

period of 24 years from 1992 to 2015 at a spatial resolution of 300 m (ESA, 2017, <http://maps.elie.ucl.ac.be/CCI/viewer/>). These maps describe the terrestrial surface of the Earth in 37 original land cover (LC) classes based on the United Nations Land Cover Classification System (UN-LCCS) (Di Gregorio, 2005). These data were developed by combining the global daily surface reflectance of 5 different observation systems, and the data accuracy was evaluated at a global scale (ESA, 2017). All datasets were transformed and re-projected to the same projection system and resolution ( $0.5^\circ \times 0.5^\circ$ ). The in situ data sampled from the atmosphere at the Mauna Loa Observatory (Keeling et al., 2005) in Hawaii was utilized as the atmospheric CO<sub>2</sub> concentration data used during the period of 1958–2015. The data before 1958 were mainly from the IS92a annual global CO<sub>2</sub> concentration dataset, and this dataset was obtained by spline fitting ice core data to the Mauna Loa sample data (Enting et al., 1994).

The global monthly means of atmospheric N<sub>2</sub>O concentration used in this study were obtained from the combined Global Monitoring Division (GMD) N<sub>2</sub>O data set (<https://www.esrl.noaa.gov/gmd/hats/combined/N2O.html>). The data set was developed by incorporating all of the monthly mean measurements of halocarbons and other atmospheric trace species (HATS) collected by several programs (e.g., old HATS flask instruments, current HATS flask instruments (OTTO), the Carbon Cycle Greenhouse Gases (CCGG) flask instruments (MAGIC), in situ HATS measurements (RITS program and CATS programs)), and they were calculated by taking the weighted averages of co-located measurements from background NOAA/ESRL GMD air measurement programs.

### 2.2. Model description

TRIPLEX-GHG is a process-based model that is primarily based on the integrated biosphere simulator (IBIS) (Foley et al., 1996) and the denitrification decomposition (DNDC) model (Li et al., 2000). The TRIPLEX-GHG model consists of six basic submodules: a land surface submodule, a vegetation dynamic submodule, a plant phenology submodule, a soil biogeochemical submodule, a methane (CH<sub>4</sub>) submodule (Zhu et al., 2014, 2015) and a N<sub>2</sub>O submodule (Zhang et al., 2017). These submodels are coupled to estimate the fluxes and pool size of carbon, nitrogen, water and three main greenhouse gases (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O) in terrestrial ecosystems at spatial scales ranging from site to regional and global scales, and time steps from hourly to yearly. The first five submodules mainly create the atmosphere-vegetation-soil system through representing energy-water exchange processes, vegetation dynamics, canopy physiology, and C and N flows for each plant functional types (PFTs) (Foley et al., 1996; Kucharik et al., 2000; Liu et al., 2005; Zhu et al., 2014). For each PFT, model balances carbon between surface and belowground carbon litter pools derived from litterfall, and soil organic matter pools of differing decomposability following Parton et al. (1987) and Verberne et al. (1990). The dynamics of the soil N pool are calculated based on the C:N ratio specified by PFT (Kucharik et al., 2000). Soil mineral N is considered to be the primary indicator of N availability. It was regulated by the decomposition of belowground litter pools, the plant uptake determined from N demand and N storage, ammonia volatilization, leaching and microbial dynamics (Foley et al., 1996; Kucharik et al., 2000). It also has feedbacks by limiting both the ecosystem C flux and the C:N ratios of the different fluxes associated with growth respiration and decomposition (Liu et al., 2005). Detailed information that describe the first five submodules could be found in the supplemental material and the key equations related to carbon balance are shown in Table S1. In the N<sub>2</sub>O submodule, the major biochemical processes that regulate N<sub>2</sub>O formation include nitrification and denitrification (Zhang et al., 2017). The decomposition process forms the linkage between N<sub>2</sub>O production and consumption pathways and soil C and N cycles in the biogeochemistry submodule. Organic C is either oxidized to CO<sub>2</sub> through microbial respiration or transferred to soluble carbon or other carbon substrates. Organic N is mineralized to

ammonium ( $\text{NH}_4^+$ ), which is then nitrified to nitrate ( $\text{NO}_3^-$ ), and these nutrients are important nutrients for nitrifiers and denitrifiers. In our model, part of the  $\text{N}_2\text{O}$  is produced through the process of biological oxidation of  $\text{NH}_4^+$  to  $\text{NO}_2^-$  and  $\text{NO}_3^-$  (nitrification pathway). The residual  $\text{N}_2\text{O}$  was produced from denitrification, which is designed to be a “chain reaction” process: the reduction of nitrate to forms nitrous oxide and molecular nitrogen. Production of  $\text{N}_2\text{O}$  by denitrification occurs when bacteria that are capable of denitrification colonize a location where oxygen is essentially absent and water, nitrate and decomposed organic compounds are present. When modeling these two microbe-based processes, the microbial activities of nitrifying and denitrifying bacteria are explicitly included based on the Michaelis–Menten equations (Michaelis and Menten, 1913), and the “anaerobic balloon” concept (Li et al., 2000) is used to regulate the rates of the allocation of substrates (e.g., DOC,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$ ) to both processes. More detailed information can be found in Zhang et al. (2017).

### 2.3. Model validation and simulations

The model has been calibrated at a global scale using daily data, and was found to perform reasonably well based on validation results in our previous study (totally 81 sites from natural forests and grasslands where little or no anthropogenic disturbances have taken place) (Zhang et al., 2017). Specifically, considering that the nitrification and denitrification processes were first coupled into the model, it is necessary to calibrate the key model parameters related to nitrification and denitrification to increase model reliability to simulate  $\text{N}_2\text{O}$  fluxes. Therefore, prior to calibration, the most sensitive parameter which strongly affect the  $\text{N}_2\text{O}$  fluxes outputs was identified for site-specific studies. Sensitivity analysis for selected 23 parameters was carried out and the maximum nitrification rate coefficient ( $\text{COE}_{\text{NR}}$ ) was found to be the most sensitive one. The optimized value of this parameter was obtained using parameter fitting for 29 calibration sites. Based on biome-type forest regions (tropical forests, temperate forests and boreal forests) and grasslands, an average parameter value for all data collection sites in that region was calculated (Table 1). To support the result of model validation, the simulation of the primary factors (soil temperature and water-filled porespace (WFPS)) was also tested by comparing the measurements for some sites where data are available. The seasonal variations and magnitudes of simulations were good overall, and annual observation and simulation data were highly correlated ( $R^2 = 0.75$ ). In this study, simulations were conducted with the parameter at the global level.

To qualify the effect of climate variability and land cover transition on  $\text{N}_2\text{O}$  fluxes, the potential effects of atmospheric deposition and land management practices were excluded from the model simulations. The model simulation went through an initial 300-year spin-up procedure. The initial 300-year spin-up was driven with multi-year (between 1901 and 1920) averaged historical meteorological data to achieve a relative equilibrium state in the carbon pools before analysis. For reaching soil carbon equilibrium, the model has an internal speed-up process during the soil spin-up period. After spin-up, the model was run starting from 1901 using daily climatological data from CRUNCEP and atmospheric

**Table 1**

Parameter (the maximum nitrification coefficient,  $\text{COE}_{\text{NR}}$ ) based on the biome type of observation sites for global forests and grasslands  $\text{N}_2\text{O}$  emissions modeling.

Biome	$\text{COE}_{\text{NR}}$	SEM	Site numbers	Data records
Boreal forests	0.09	0.0136	4	126
Temperate forests	0.04	0.0124	15	665
Tropical forests	0.009	0.0018	4	200
Grasslands	0.03	0.0047	6	333

SEM: standard error of the mean.

$\text{CO}_2$  data based on ice core and atmospheric measurements for transient simulations (Keeling et al., 2005). The global vegetation map used for model initial simulation from 1901 to 1991 was generated from the GlobCover 2009 land cover map (original spatial resolution: 300 m) (Bontemps et al., 2010), and the ESA – CCI – LC (annual land cover map available during the period of 1992–2015) was used for subsequent simulations. Only result of 1992–2015 were extracted for analysis.

### 2.4. Trend and correlation analysis

The total  $\text{N}_2\text{O}$  emission (T) and the area-weighted average  $\text{N}_2\text{O}$  flux ( $F_s$ ) are calculated as follows:

$$T = \sum_{j=1}^n F_j \cdot S_j$$

$$F_s = \frac{T}{\sum_{j=1}^n S_j}$$

where n is the total number of grids of forests or grasslands;  $F_j$  and  $S_j$  are the  $\text{N}_2\text{O}$  flux and the area of the jth grid, respectively. The vegetation type for an individual grid in each year is classified based on the annual ESA – CCI – LC maps.

The trends of  $\text{N}_2\text{O}$  emissions for individual grid cells were identified using the Mann–Kendall test. The Mann–Kendall test is a nonparametric technique (Mann, 1945; Kendall, 1948) that is widely used in hydrology and climatology. The Mann–Kendall statistic indicates the direction and magnitude of the trend in simulated natural  $\text{N}_2\text{O}$  emissions. In the Mann–Kendall trend test for a set of elements, n is the total number of elements; here we used the total number of annual data. The Mann–Kendall rank test statistic S is given by:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^n \text{Sgn}(X_j - X_k)$$

$$\text{Sgn}(X_j - X_k) = \begin{cases} 1 & (X_j - X_k) > 0 \\ 0 & (X_j - X_k) = 0 \\ -1 & (X_j - X_k) < 0 \end{cases}$$

where n is the total number of elements;  $X_j$  and  $X_k$  are the jth and kth elements ( $j \leq n$ , and  $k \neq j$ ).

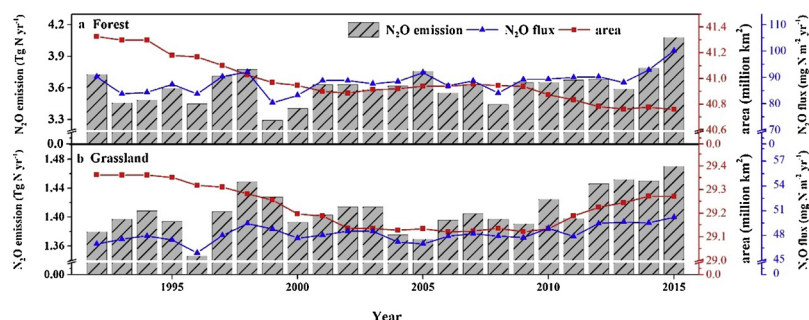
In the absence of any trend ( $H_0$  or null hypothesis), the function of S can be assumed to have a normal distribution, with the expected variance  $\text{var}(S)$  given by:

$$\text{var}(S) = \sigma^2 = n \cdot (n - 1) \cdot (2n + 5) / 18$$

The standard normal distribution Z is given by:

$$Z = \begin{cases} (S - 1) / \sqrt{\text{var}(S)} & S > 0 \\ 0 & S = 0 \\ (S + 1) / \sqrt{\text{var}(S)} & S < 0 \end{cases}$$

The distribution of the test statistic Z is compared with a standard normal distribution at a certain level of significance, and the significance level is set to be 0.05 in this study. Therefore, the no trend or  $H_0$  hypothesis is rejected for high values of  $|Z| \geq 1.96$ , and the trend is considered increasing when Z is greater than 0. We mapped the spatial pattern of this statistic for modeled  $\text{N}_2\text{O}$  emissions to identify significant trends. Furthermore, Spearman correlation analysis was used to estimate the correlation between the  $\text{N}_2\text{O}$  fluxes and the model-driven data over the time series (i.e., the input climate data). All statistical analyses were performed using the open-source software R version 3.2.0 (R Foundation for Statistical Computing, Vienna, Austria).



**Fig. 1.** Interannual variations of  $\text{N}_2\text{O}$  emissions,  $\text{N}_2\text{O}$  fluxes and areas of global forests (a) and grasslands (b). The  $\text{N}_2\text{O}$  emission means the total emission calculated by summing the products of  $\text{N}_2\text{O}$  flux and area; the  $\text{N}_2\text{O}$  flux means the per unit area  $\text{N}_2\text{O}$  emission.

### 3. Results

#### 3.1. Forest and grassland $\text{N}_2\text{O}$ emission budgets at global, biome and continental scales

As shown in Fig. 1, the estimated total global forest areas decreased from 41.76 million  $\text{km}^2$  to 41.32 million  $\text{km}^2$ . The global area-weighted mean  $\text{N}_2\text{O}$  flux from forests from 1992 to 2015 was  $88.3 \pm 4.0 \text{ mg N m}^{-2} \text{ yr}^{-1}$ , ranging from  $80.3 \text{ mg N m}^{-2} \text{ yr}^{-1}$  (1999) to  $100.1 \text{ mg N m}^{-2} \text{ yr}^{-1}$  (2015). The estimated average total  $\text{N}_2\text{O}$  emissions from forests from 1992 to 2015 was  $3.62 \pm 0.16 \text{ Tg N yr}^{-1}$ , ranging from  $3.29 \text{ Tg N yr}^{-1}$  (1999) to  $4.08 \text{ Tg N yr}^{-1}$  (2015).

The estimated total global grassland areas first decreased until 2004 and then increased from 2004 to 2015, with the average increase of  $29.2 \pm 0.09 \text{ million km}^2 \text{ yr}^{-1}$ . The global area-weighted mean  $\text{N}_2\text{O}$  flux from grasslands from 1992 to 2015 was  $48.2 \pm 1.0 \text{ mg N m}^{-2} \text{ yr}^{-1}$ , ranging from  $45.9 \text{ mg N m}^{-2} \text{ yr}^{-1}$  (1996) to  $50.2 \text{ mg N m}^{-2} \text{ yr}^{-1}$  (2015). The estimated total  $\text{N}_2\text{O}$  emissions from grasslands from 1992 to 2015 was approximately  $1.40 \pm 0.03 \text{ Tg N yr}^{-1}$ , ranging from  $1.35 \text{ Tg N yr}^{-1}$  (1996) to  $1.47 \text{ Tg N yr}^{-1}$  (2015). Moreover,  $\text{N}_2\text{O}$  emissions showed slightly increasing trends for both forests (mean increase of  $9.9 \text{ Gg N yr}^{-1}$ ) and grasslands (mean increase of  $2.1 \text{ Gg N yr}^{-1}$ ) over the study period. Approximately 83.9% and 74% of the total emissions were from forests and grasslands in tropical regions, respectively. Boreal regions contributed a smaller proportion of  $\text{N}_2\text{O}$  emissions than contributed by areas in tropical regions or other biomes (Table 2, Figs. S1, S2). The modeled total budget of annual  $\text{N}_2\text{O}$  emissions from global forests and grasslands can be found in Table S2.

At the continental scale, South America showed the highest  $\text{N}_2\text{O}$  emissions from both forest and grassland ecosystems, which is caused by the large  $\text{N}_2\text{O}$  fluxes compared to other continents (Table 3). Africa and South America acted as the largest forest and grassland  $\text{N}_2\text{O}$  sources as they had the highest  $\text{N}_2\text{O}$  fluxes and the largest natural forest and grassland areas, suggesting that Africa and South America play major roles in the global  $\text{N}_2\text{O}$  budget at the continental scale.

#### 3.2. Spatial distribution and trends of $\text{N}_2\text{O}$ fluxes

Fig. 2a, b shows the spatial distribution of  $\text{N}_2\text{O}$  fluxes in forests and grasslands, respectively. Rate of  $\text{N}_2\text{O}$  released greater than  $550 \text{ mg N m}^{-2} \text{ yr}^{-1}$  were found in tropical and subtropical forest regions and

savannas due to the greater availability of substrate and favorable climate conditions for nitrifiers and denitrifiers. However, the large boreal forest areas in northern high-latitude regions were less-substantial  $\text{N}_2\text{O}$  emission sources ( $< 20 \text{ mg N m}^{-2} \text{ yr}^{-1}$ ). Fig. 2c, d shows the results of the Mann-Kendall trend test for  $\text{N}_2\text{O}$  fluxes in forests and grasslands. Most regions showed increasing simulated  $\text{N}_2\text{O}$  fluxes, and significant increases were found in tropical forests, such as the Amazon plain and Central Africa, subtropical forests in southeastern China and southwest Europe and boreal forests in Eastern and Central Russia. Some areas (grid cells) that were mainly distributed along the Eastern Andes and Southeast Asia showed significant decreasing trends. Grasslands located in northern North America, Pampas in Africa, Inner Mongolia in China and southwest Australia showed significant decreases in simulated  $\text{N}_2\text{O}$  fluxes, while the majority of grid cells in Africa showed significant increasing trends.

#### 3.3. Seasonal trends in $\text{N}_2\text{O}$ fluxes and emissions in global forests and grasslands

Fig. 3 shows the seasonal variations of  $\text{N}_2\text{O}$  fluxes along a zonal gradient and total  $\text{N}_2\text{O}$  emissions. There are distinct seasonal cycles in the  $\text{N}_2\text{O}$  emissions from natural forests and grasslands. Forests emissions from Northern Hemisphere and Southern Hemisphere both exhibited one seasonal peak, with the peak emissions occurring in July ( $0.284 \pm 0.016 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ) and December ( $0.197 \pm 0.013 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ), respectively. It is obvious that the seasonal variations amplitudes in simulated global forest  $\text{N}_2\text{O}$  emissions from Northern Hemisphere were larger than that from Southern Hemisphere. For grasslands, the seasonal variation of emissions showed an opposite trend. For example, the highest emission occurred in July ( $0.107 \pm 0.005 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ) and lowest in December ( $0.037 \pm 0.004 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ) from Northern Hemisphere, and the highest emission occurred in January ( $0.085 \pm 0.005 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ) and lowest in June ( $0.032 \pm 0.002 \text{ Tg N m}^{-2} \text{ month}^{-1}$ ) from Southern Hemisphere. The increases in solar radiation and growing season length from north to south could be responsible for the existing pattern. Although, Northern Hemisphere  $\text{N}_2\text{O}$  emissions from both grasslands and forests were higher than that in Southern Hemisphere, it is based on a fact that the Northern land area is larger than the south. For forests and grasslands, the average  $\text{N}_2\text{O}$  fluxes along the altitudinal gradient first spread

**Table 2**

Average percentages of total  $\text{N}_2\text{O}$  emissions and total areas of forests and grasslands in different biomes from 1992 to 2015 (the values are denoted as mean  $\pm$  SD (standard deviation)).

Percentage of total amount	Ecosystem type	Boreal region	North temperate region	Tropical region	South temperate region
$\text{N}_2\text{O}$ emissions	Forest	$1\% \pm 0.15\%$	$13.8\% \pm 1\%$	$83.9\% \pm 1\%$	$1.3\% \pm 0.08\%$
	Grassland	$0.9\% \pm 0.15\%$	$18\% \pm 0.7\%$	$74\% \pm 0.7\%$	$7.1\% \pm 0.3\%$
Total area	Forest	$15.6\% \pm 0.09\%$	$33.3\% \pm 0.17\%$	$49.4\% \pm 0.26\%$	$1.7\% \pm 0.01\%$
	Grassland	$8.4\% \pm 0.15\%$	$34.5\% \pm 0.14\%$	$50.1\% \pm 0.26\%$	$7\% \pm 0.03\%$



**Table 3**

Average N<sub>2</sub>O fluxes, areas and emissions from forests and grasslands on different continents from 1992 to 2015 as simulated by the TRIPLEX-GHG model (the values are denoted as mean  $\pm$  SD (standard deviation)).

Forest				Grassland		
Continents	N <sub>2</sub> O emission (Tg N yr <sup>-1</sup> )	Area (million km <sup>2</sup> )	N <sub>2</sub> O flux (mg N m <sup>-2</sup> yr <sup>-1</sup> )	N <sub>2</sub> O emission (Tg N yr <sup>-1</sup> )	Area (million km <sup>2</sup> )	N <sub>2</sub> O flux (mg N m <sup>-2</sup> yr <sup>-1</sup> )
Asia	0.71 $\pm$ 0.037	12.7 $\pm$ 0.05	60.0 $\pm$ 3.0	0.22 $\pm$ 0.01	8.1 $\pm$ 0.04	26.9 $\pm$ 1.4
North America	0.36 $\pm$ 0.02	7.7 $\pm$ 0.03	46.8 $\pm$ 2.5	0.20 $\pm$ 0.007	5.33 $\pm$ 0.015	37.6 $\pm$ 1.23
Europe	0.053 $\pm$ 0.006	3.5 $\pm$ 0.012	15.13 $\pm$ 1.63	0.025 $\pm$ 0.002	1.18 $\pm$ 0.02	20.6 $\pm$ 1.6
Africa	1.18 $\pm$ 0.036	7.03 $\pm$ 0.04	167.6 $\pm$ 4.46	0.40 $\pm$ 0.008	7.0 $\pm$ 0.13	57.6 $\pm$ 1.5
South America	1.26 $\pm$ 0.11	7.88 $\pm$ 0.16	160.3 $\pm$ 13.9	0.45 $\pm$ 0.016	4.42 $\pm$ 0.04	102.55 $\pm$ 3.2
Oceania	0.054 $\pm$ 0.004	0.72 $\pm$ 0.0023	74.8 $\pm$ 5.2	0.11 $\pm$ 0.009	3.18 $\pm$ 0.02	34.8 $\pm$ 2.7

southwards and then northwards throughout the year. The relatively high level of monthly N<sub>2</sub>O fluxes from forests (generally more than 20 mg N m<sup>-2</sup> month<sup>-1</sup>) occurred between 30°N and 40°N during summer, while in grasslands, they occurred near 20°N in Spring.

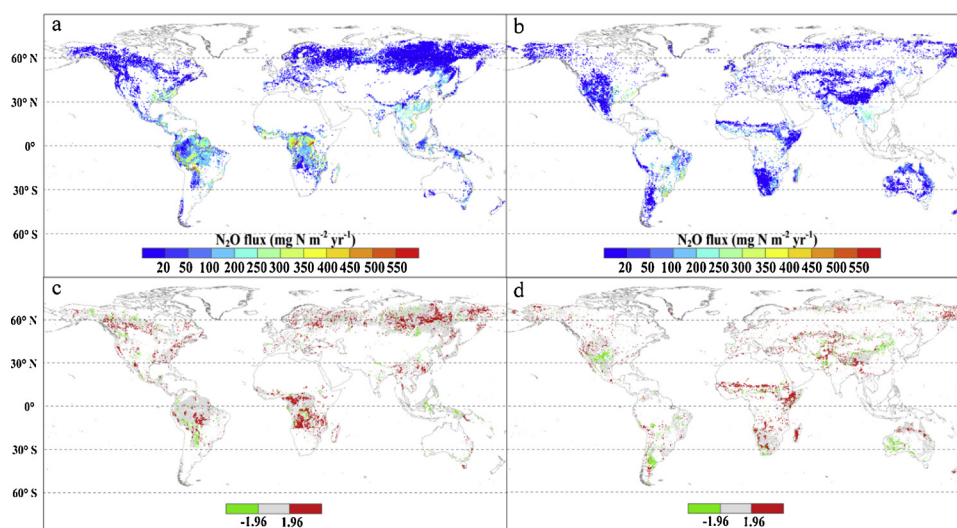
### 3.4. The correlation between climate forcings and global N<sub>2</sub>O fluxes from global forests and grasslands

Fig. 4 shows the Spearman correlations between global N<sub>2</sub>O fluxes and precipitation (Fig. 4a, b) and temperature (Fig. 4c, d). Compared with air temperature, precipitation is the dominant controller that regulates the N<sub>2</sub>O fluxes from the majority of forests and grasslands. For global forests and grasslands, the total number of grids where the N<sub>2</sub>O flux is significant correlated to the precipitation and temperature are 16,385 and 6369, which accounted for approximately 55.3 and 21.5 percent of total grids, respectively. And about 25.3 percent of grids showed the correlation coefficient between N<sub>2</sub>O flux and precipitation was bigger than 0.5 with significance. In most boreal forest ecosystems, N<sub>2</sub>O fluxes were positively correlated with precipitation and specific humidity, while they were not significantly correlated ( $P > 0.05$ ) with other climate variables (Figure S3). For tropical forest regions, N<sub>2</sub>O fluxes were significantly negatively correlated with precipitation in the majority of tropical forest regions areas, except for some scattered regions in the Congo Basin that showed positive correlations.

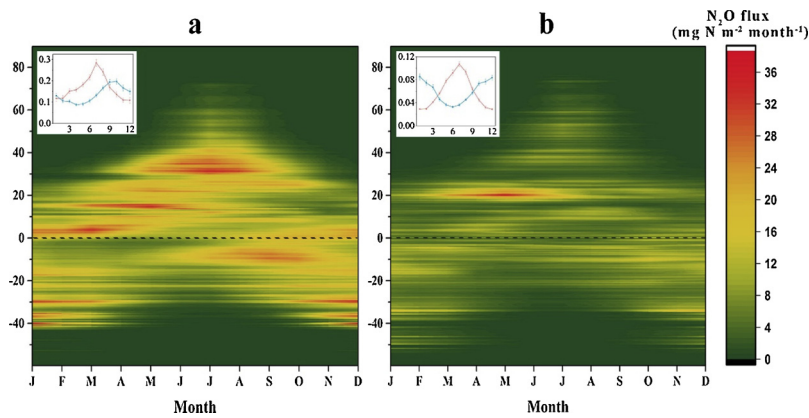
In the majority of grassland ecosystems, precipitation was positively correlated with N<sub>2</sub>O fluxes. Precipitation was positively correlated with the N<sub>2</sub>O fluxes in most of the temperate grassland regions in the Southern Hemisphere, except for those in Madagascar, Africa. Furthermore, temperature and precipitation showed opposite effects on the N<sub>2</sub>O fluxes from grasslands in Australia, while they showed consistent effects in Eastern Russia and the Qinghai-Tibet Plateau in China.

### 3.5. The relationship between N<sub>2</sub>O emissions and El Niño–southern oscillation (ENSO)

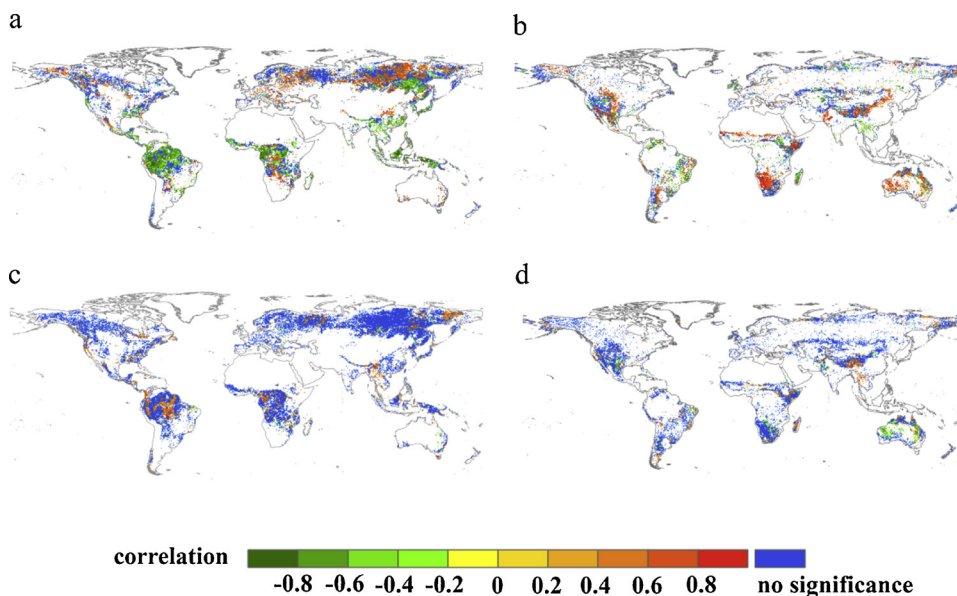
Fig. 5 shows the spatial distributions of N<sub>2</sub>O fluxes, precipitation and temperature increases (the value in 1998 minus that in 1999) for tropical forests. The N<sub>2</sub>O flux in most tropical forests appeared to decrease at various degrees. Specifically, between 1998 and 1999, there were reductions of more than 300 mg N m<sup>-2</sup> yr<sup>-1</sup> in the N<sub>2</sub>O fluxes in the Orinoco plain and northeast Amazon Plain in South America. To illustrate the relationship between tropical N<sub>2</sub>O emissions and the ENSO, the monthly mean anomaly fluxes of N<sub>2</sub>O in tropical forests and grassland ecosystems during the experimental periods were performed according to La Niña and El Niño events. According to Fig. 6, the monthly mean anomaly fluxes of N<sub>2</sub>O tend to be greater than zero during strong and very strong La Niña events periods for tropical forests, which indicates an increase of N<sub>2</sub>O emissions comparing the average level. The monthly mean anomaly fluxes of N<sub>2</sub>O tend to be less than zero during strong and very strong El Niño events, which indicates a decrease of N<sub>2</sub>O emissions. For grassland ecosystem, the monthly mean anomaly fluxes of N<sub>2</sub>O during La Niña and El Niño events varies without regularity. Fig. 7 shows the differences in the area-weighted mean monthly N<sub>2</sub>O fluxes from tropical forests and grasslands between La Niña or El Niño months and neutral months. The responses of N<sub>2</sub>O release from tropical forests to El Niño events exhibit the opposite pattern (Fig. 7a). The N<sub>2</sub>O fluxes during El Niño months are larger than those during neutral months. For grasslands in the tropics, wet months (generally from May to October) during El Niño/La Niña years showed relatively higher/lower average N<sub>2</sub>O fluxes, while dry months (generally from November to the following April) during El Niño/La Niña years showed relatively lower/higher average N<sub>2</sub>O fluxes than those in the corresponding months in neutral years (Fig. 7b).



**Fig. 2.** Spatial patterns of the average N<sub>2</sub>O fluxes for forests (a) and grasslands (b); Mann-Kendall statistics in forests (c) and grasslands (d) simulated for 1992–2015.



**Fig. 3.** Variations of area-weighted average  $\text{N}_2\text{O}$  fluxes and total emissions ( $\text{Tg N month}^{-1}$ , graphs at the left upper corner) from forests (a) and grasslands (b) along with the latitudinal gradient (by  $1^\circ$ ) at a monthly step. For the graphs at the left upper corner, the red line represents  $\text{N}_2\text{O}$  emissions from the Northern Hemisphere, and the blue line represents  $\text{N}_2\text{O}$  emissions from the Southern Hemisphere (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).



**Fig. 4.** Correlations between  $\text{N}_2\text{O}$  fluxes and precipitation in forests (a) and grasslands (b), and between  $\text{N}_2\text{O}$  fluxes and temperature in forests (c) and grasslands (d). Non-significant correlations are shown in blue (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

### 3.6. The relationship between soil $\text{N}_2\text{O}$ emissions from forest and grasslands and atmospheric $\text{N}_2\text{O}$ concentrations

Overall, the correlation between soil  $\text{N}_2\text{O}$  emissions and atmospheric  $\text{N}_2\text{O}$  concentrations was low (Figure S4,  $R^2 = 0.26$ ) when the sum of the emissions from forests and grasslands was considered which implies that approximately 25.9% of total observed annual  $\text{N}_2\text{O}$  increases in the atmosphere can be explained by these natural emissions (forests and grasslands). The significant correlations were mainly attributed to strong  $\text{N}_2\text{O}$  concentration variations for specific months. From June to November, soil  $\text{N}_2\text{O}$  emissions from forests are significantly positively correlated with the atmospheric  $\text{N}_2\text{O}$  concentrations, as the  $\text{N}_2\text{O}$  emissions from forests are larger from June to November than those in other months (Figs. 3a, 5). For grassland ecosystems, soil  $\text{N}_2\text{O}$  emissions have significant positive correlation to the atmospheric  $\text{N}_2\text{O}$  concentrations in January, September, October and November (Table 4), however, it may be caused by the co-correlated with positive relationship between concentration and emissions in forest, based on the fact the overall grassland emissions are much smaller than forest emissions.

### 3.7. Sensitivity analysis of spatial estimation

Sensitivity experiments have been conducted for different climate variables, including the precipitation and air temperature. The value of a single input parameter is changed ( $\pm 25\%$  for precipitation and  $\pm$

$0.5^\circ\text{C}$  for air temperature) relative to its original value, with other parameters held fixed. In addition, we applied the simplified Most Significant Factor (MSF) method to estimate the uncertainties induced by parameters that are fluctuated in a certain range (Li et al., 1996; Giltrap et al., 2010). The MSF method involves taking the extreme values of the factor(s) producing most of the variation in the model predictions and can be used in most regional simulations with low computationally expensive. The key parameter  $\text{COE}_{\text{NR}}$  was only conducted the MSF method, since it turns out from our model study (Zhang et al., 2017) to be the most sensitive parameter in controlling  $\text{N}_2\text{O}$  emissions to the atmosphere and it differs in biome regions when conducting the regional simulation.

The  $\text{N}_2\text{O}$  emission amounts resulting from each run are then compared with the result from the standard run (SS) for different biome regions (Table 5). For tropical and temperate forests, the climate variables have a significant effect on global  $\text{N}_2\text{O}$  emissions. The total emissions have been changed more than 10% for changing 25% of precipitation and changing  $0.5^\circ\text{C}$  of air temperature. However climate variables seem to have relatively weak influence on  $\text{N}_2\text{O}$  emissions from boreal regions. The precipitation variation has opposite effect on tropical forests  $\text{N}_2\text{O}$  emissions compared to air temperature, for example, the emissions increasing by  $0.53 \text{ Tg N yr}^{-1}$  when the precipitation is decreased by 25%. Air temperature performed positive effect on all regions of global forests and grasslands, with about 26% increase and 53% decrease in total  $\text{N}_2\text{O}$  emissions when the air temperature is increase and decrease  $0.5^\circ\text{C}$ , respectively. Based on the MS and NS

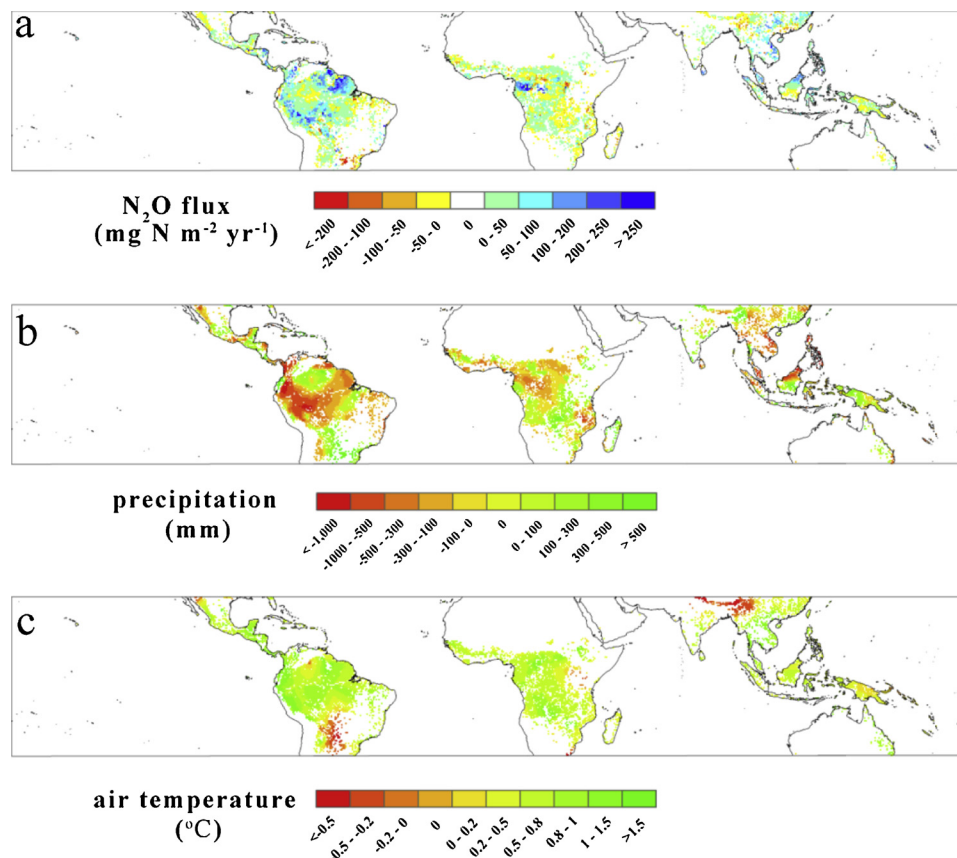


Fig. 5. Spatial distributions of N<sub>2</sub>O fluxes, precipitation and temperature increases (the value in 1998 minus that in 1999) for tropical forests.

scenarios (Table 5), the extreme COE<sub>NR</sub> values for every biome regions produced a range of N<sub>2</sub>O emission predictions (about -14.6%~+11.9%). Furthermore, the variation of COE<sub>NR</sub> may not change the trends of N<sub>2</sub>O emissions during the period of 1992–2015.

## 4. Discussion

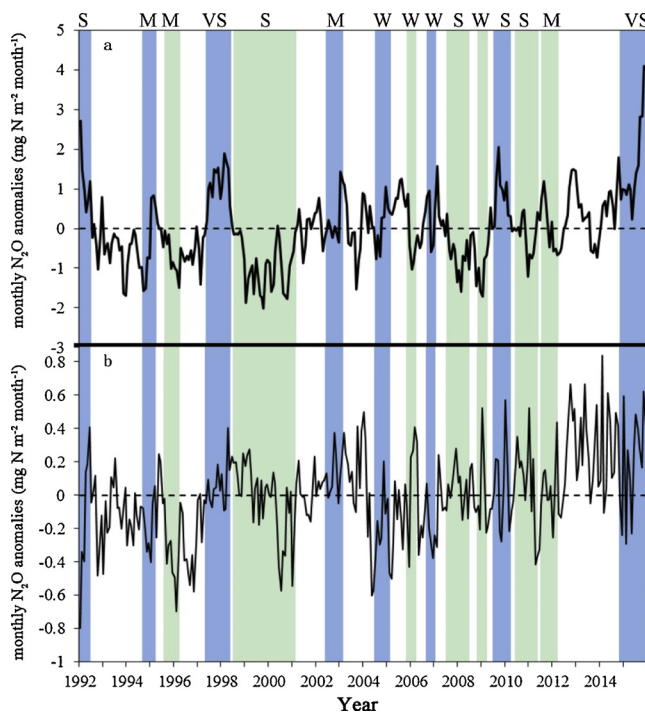
### 4.1. Comparisons with previous studies

The total predicted N<sub>2</sub>O emissions of 3.62 Tg N per year from forests and 1.41 Tg N per year from grasslands during the study period are both within the range of previously reported values. Tian et al. (2013) reported average N<sub>2</sub>O emissions of 4.28 Tg N per year from global forests and 3.64 Tg N per year from global grasslands over 1981–2010. The lower N<sub>2</sub>O estimates of our study may be due to the effects of additional global change factors such as tropospheric O<sub>3</sub>, nitrogen deposition and nitrogen fertilizer use, which are not included in the model in this study, and these factors were reported to increase N<sub>2</sub>O emissions via their influence on soil C and N balance (Gomez-Casanovas et al., 2016; Kanerva et al., 2008; Shcherbak et al., 2014; Xu et al., 2008). Moreover, Xu et al. (2008) also reported global N<sub>2</sub>O emission rates of 6.99 Tg N per year for forests and 4.49 Tg N per year for grasslands from 2000 to 2008 by combining an empirical climate-driven soil respiration model (Raich et al., 2002) (driven by air temperature and precipitation) with the linear functions of N<sub>2</sub>O and CO<sub>2</sub> fluxes, which are obtained from a meta-analysis. The N<sub>2</sub>O emissions estimated by Xu et al. (2008) are much higher than those estimated in this study, which may be due to overestimating of soil respiration and ignoring the effects of other important factors (e.g., soil pH) on N<sub>2</sub>O emissions (Xu et al., 2008). Zhuang et al. (2012) reported values of 1.3 Tg N per year from forests and 1.31 Tg N per year from grasslands in 2000, and these values were extrapolated from field measurements by using an artificial neural

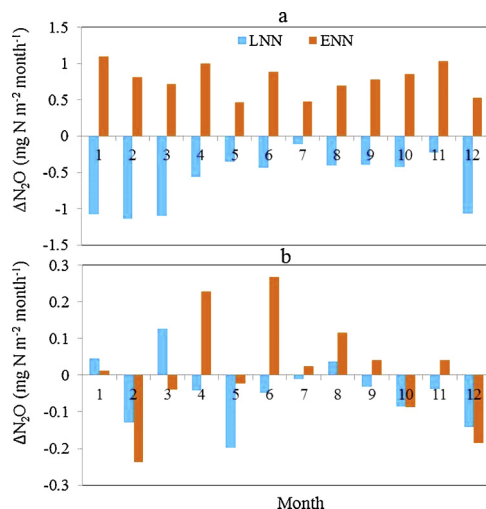
network approach; the differences in the N<sub>2</sub>O emissions from forests between the two studies may be caused by the differences in LC area estimates. In our study, the forest area was estimated to be 40.95 million km<sup>2</sup> (ESA-CCI-LC), and this estimated value is similar to the value estimated by Keenan et al. (2015) (40.55 million km<sup>2</sup> in 2000), while the forest area estimated by Zhuang et al. (2012) (Land Cover Type Yearly L3 Global CMG, MCD12C1; 26.03 million km<sup>2</sup> in 2000) is much smaller than that estimated in this study. The N<sub>2</sub>O emissions from tropical forests were estimated to be within the range of 1.17–3.55 Tg N yr<sup>-1</sup> (Breuer et al., 2000; Matson and Vitousek, 1990; Potter et al., 1996; Stehfest and Bouwman, 2006; Werner et al., 2007) during the 1990s, and the N<sub>2</sub>O emissions from tropical forests estimated by our model (2.98 Tg N yr<sup>-1</sup>) were within this range.

In addition, the global spatial patterns of N<sub>2</sub>O fluxes obtained in this study are also consistent with the results of other studies (Saikawa et al., 2013; Xu et al., 2012; Zhuang et al., 2012). In general, boreal regions showed relatively low N<sub>2</sub>O emissions (< 40 mg N m<sup>-2</sup> yr<sup>-1</sup>), while tropical regions (especially in rainforest area) showed relatively higher emissions (usually more than 200 mg N m<sup>-2</sup> yr<sup>-1</sup>). However, great uncertainty still exists in the estimation of N<sub>2</sub>O fluxes in tropical regions (Tian et al., 2016). The N<sub>2</sub>O fluxes estimated by this study in tropical regions showed significant spatial heterogeneity, and the estimations of N<sub>2</sub>O fluxes in rainforest regions that were close to the equator could reach 685 mg N m<sup>-2</sup> yr<sup>-1</sup>; however, some areas showed extremely low N<sub>2</sub>O fluxes (near zero). Conversely, in the study by Xu et al. (2012), the differences in N<sub>2</sub>O fluxes from tropical regions were small; that is, the majority of places showed high N<sub>2</sub>O emission levels. The results of Saikawa et al. (2013) showed that the N<sub>2</sub>O emissions from southern Asia and Southeast Asia play major roles in the global N<sub>2</sub>O budget, while in this study, the forests in Africa and South America that were close to the equator were the main sources of N<sub>2</sub>O emissions. This finding indicates that tropical regions have become the areas with





**Fig. 6.** Monthly mean anomaly fluxes of  $N_2O$  (total monthly emissions/total area) in tropical forest (a) and savanna (b) ecosystems between 1992 and 2015 ( $mg\ N\ m^{-2}\ month^{-1}$ ). The green and blue areas represent the periods of La Niña and El Niño events respectively (S: strong; M: moderate; VS: very strong; W: weak, [http://origin.cpc.ncep.noaa.gov/products/analysis\\_monitoring/ensostuff/ONI\\_v5.php](http://origin.cpc.ncep.noaa.gov/products/analysis_monitoring/ensostuff/ONI_v5.php)) (Huang et al., 2017) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).



**Fig. 7.** Differences in the area-weighted mean monthly  $N_2O$  fluxes from tropical forests (a) and grasslands (b) between La Niña or El Niño months and neutral months. LNN and ENN represent the values derived from the area-weighted monthly mean  $N_2O$  fluxes from La Niña and El Niño months minus the area-weighted monthly mean  $N_2O$  fluxes from neutral months, respectively. The periods of La Niña and El Niño event are based on [http://origin.cpc.ncep.noaa.gov/products/analysis\\_monitoring/ensostuff/ONI\\_v5.php](http://origin.cpc.ncep.noaa.gov/products/analysis_monitoring/ensostuff/ONI_v5.php). The mean and standard deviations of area-weighted monthly  $N_2O$  fluxes from forests and grasslands in neutral month, El Niño month and La Niña month can be found in Table S3.

high uncertainties in  $N_2O$  flux estimates. On the one hand, this fact is probably due to the effects of the different responses of climate factors (particularly under high rainfall and high temperature conditions) on  $N_2O$  emissions in different models. On the other hand, tropical regions have unique water thermal environment conditions and the geographical positions that result in the frequent occurrence of extreme climate events, which may be the cause of the large spatial heterogeneity of the  $N_2O$  emissions from tropical regions.

#### 4.2. The relationship between $N_2O$ emissions and El Niño–southern oscillation (ENSO)

The anomalous climate patterns in tropical regions were mainly induced by La Niña/El Niño events, and such anomalies from normal surface temperatures caused by La Niña/El Niño events can have large-scale impacts not only on ocean processes but also on global weather and climate. The mature La Niña from November 1998 to early 2001 was reported by Schwing et al. (2002), and this study revealed that the  $N_2O$  fluxes in the tropics decreased during this period. Similarly, the decreases in the total emissions in 1996 and 2008 were also preceded by sharp decreases in the  $N_2O$  fluxes in tropical regions that were coincident with anomalous climate patterns that were induced by the recorded La Niña events during the corresponding periods (Schwing et al., 2002). For example, the  $N_2O$  flux between 1998 and 1999 has large reductions, especially in the Orinoco plain and northeast Amazon Plain in South America, it can be ascribed to the substantial increase in precipitation induced by the La Niña events and as a result of the significant negative correlation between the  $N_2O$  flux and precipitation in tropical regions. Tropical forests  $N_2O$  flux during neutral months are less than those during El Niño months and larger than those during La Niña months. However, this is in contrast to the prevailing view that El Niño/La Niña events induce  $N_2O$  decreases/increases (Huang and Gerber, 2015; Saikawa et al., 2013; Thompson et al., 2013), it may be due to the different model performance under the high level of precipitation. In this study, excessive precipitation induced the decrease of  $N_2O$  fluxes for tropical regions. In addition, for grasslands in the tropics,  $N_2O$  fluxes during El Niño/La Niña events showed relatively higher/lower in wet months (generally from May to October) and lower/higher in dry months (generally from November to the following April) comparing the neutral months. This response may be attributed to the tropical savanna climate, which has distinct dry and wet seasons. Greenhouse gases (i.e.,  $CO_2$ ,  $N_2O$  and  $CH_4$ ) have been proven to be sensitive to extreme climate events (Zhu et al., 2017; Gurney et al., 2012; Schwalm et al., 2011); thus, extreme climate events may become comprehensive and effective indicators that reflect the relationship between climate change and global greenhouse gas emissions.

#### 4.3. The relationship between $N_2O$ emissions from forests and grasslands and the atmospheric $N_2O$ concentration

The interannual variations of  $N_2O$  emitted from natural forest and grassland soils showed slight correlations with the consistent increase in the atmospheric  $N_2O$  concentration, which implies that the increasing atmospheric  $N_2O$  concentration is mainly due to the elevation of other emission sources, such as emissions from anthropogenic, riverine and oceanic  $N_2O$  (Flückiger et al., 1999; Hu et al., 2016). Complicated processes, including gas diffusion and chemical reactions in the atmosphere (Ravishankara et al., 2009), are another reason for the low correlations between the atmospheric  $N_2O$  concentration and  $N_2O$  emissions from forests and grasslands. In addition, there is no distinct seasonal variation in the atmospheric  $N_2O$  concentration; however, we found that their correlations differed in different months. For example, soil  $N_2O$  emissions from forests during summer and autumn have positive effects on the atmospheric  $N_2O$  concentrations, while no significant correlations were found in spring and winter. This result might be related to the rapid growth rates of  $N_2O$  during summer and autumn



**Table 4**The monthly mean atmospheric N<sub>2</sub>O concentration and N<sub>2</sub>O emissions from forests or grasslands and their correlations.

Atmospheric N <sub>2</sub> O concentration (ppm)			N <sub>2</sub> O emissions from forests (Tg N month <sup>-1</sup> )			N <sub>2</sub> O emissions from grasslands (Tg N month <sup>-1</sup> )		
Month	Mean	SD	Mean	SD	R <sup>2</sup>	Mean	SD	R <sup>2</sup>
1	317.991	5.617	0.246	0.022	0.006	0.114	0.005	0.274 <sup>a</sup>
2	318.040	5.595	0.224	0.020	0.001	0.104	0.004	0.063
3	318.023	5.594	0.254	0.015	0.003	0.108	0.004	0.133
4	318.003	5.623	0.246	0.012	0.011	0.102	0.004	0.005
5	317.994	5.640	0.273	0.012	0.110	0.114	0.004	0.002
6	318.014	5.644	0.322	0.014	0.281 <sup>a</sup>	0.124	0.006	0.060
7	318.068	5.648	0.416	0.018	0.143 <sup>a</sup>	0.143	0.005	0.000
8	318.147	5.666	0.408	0.021	0.187 <sup>a</sup>	0.139	0.004	0.008
9	318.251	5.693	0.363	0.024	0.256 <sup>a</sup>	0.121	0.004	0.166 <sup>a</sup>
10	318.404	5.728	0.333	0.022	0.257 <sup>a</sup>	0.118	0.004	0.217 <sup>a</sup>
11	318.545	5.756	0.276	0.023	0.155 <sup>a</sup>	0.109	0.005	0.279 <sup>a</sup>
12	318.683	5.749	0.258	0.024	0.116 <sup>a</sup>	0.112	0.004	0.039

SD: standard deviation.

<sup>a</sup> significant correlation.

and the relatively low or negative growth rates during spring and winter. Atmospheric N<sub>2</sub>O concentrations significantly increased with the increase in soil emissions from grasslands in autumn, which is the growing season for grasses in the Southern Hemisphere. Thus, natural sources (forest and grassland ecosystems) are not the key contributors to the rising atmospheric N<sub>2</sub>O concentration.

#### 4.4. The relationship between N<sub>2</sub>O emissions and the climate factors

Additionally, the model results show that the spatial and seasonal distributions of N<sub>2</sub>O emissions are highly related to climate patterns (mainly the precipitation and temperature patterns). For most grasslands, increases in precipitation would favor the production of N<sub>2</sub>O. Previous studies indicated that N<sub>2</sub>O fluxes are sensitive to precipitation (Butterbach-Bahl et al., 2000; Li et al., 2000; Lu et al., 2008), since precipitation plays a dominant role in controlling soil moisture, which is vital to gas diffusion, denitrification and N<sub>2</sub>O emissions. Increases in soil moisture provide anaerobic conditions for N<sub>2</sub>O production and promote the decomposition of residual organic matter, enhancing the supply of nitrogen and carbon substrates for denitrification (Chen et al., 2013). In the tropics where temperature is not a limiting factor, moisture is the dominant factor for N<sub>2</sub>O emissions. However, elevated precipitation was found to be negatively correlated with N<sub>2</sub>O fluxes from tropical regions in our study, and a similar effect was reported by several other studies. Weitz et al. (2001) reported that nitrification decreases with increasing soil moisture contents and is predicted to cease at approximately 70% WFPS. Reduction of N<sub>2</sub>O to N<sub>2</sub> is expected to start at 70% WFPS and to increase rapidly with increasing soil

saturation (Weitz et al., 2001). Castaldi et al. (2013) also reported that there is a progressive reduction in the lengths dry periods in tropical forests with increases in the rainfall rates (with a peak gas flux between 30% and 35% WFPS), and these conditions are not favorable for N<sub>2</sub>O production. Similarly, decreases in N<sub>2</sub>O emissions in relation to the large increases in precipitation were also detected by the DyN-LPJ model (Xu et al., 2012) and Forest-DNDC model (Werner et al., 2007).

#### 4.5. Uncertainties

Estimations of regional and global N<sub>2</sub>O budgets have large uncertainties as a result of the high temporal and spatial variations of the input parameters within natural forests and grasslands. On one hand, COE<sub>NR</sub> turns out from our model study to be a very important parameter in controlling N<sub>2</sub>O emissions to the atmosphere. It was used to regulate the nitrification rate, therefore, the predominant sensitivity of N<sub>2</sub>O emissions to COE<sub>NR</sub> can be explained by the increase of direct N<sub>2</sub>O production from nitrification and the availability of denitrification substrates. The experiments of MSF for COE<sub>NR</sub> indicated that the variation of N<sub>2</sub>O emissions is more likely in the range of -14.6%~+11.9%. Meteorological time series such as temperature and precipitation also have their measurement errors, thus induce some uncertainties. Most importantly, they are sensitive to the N<sub>2</sub>O emissions. Temporal averaging and interpolation also introduce errors, for example, the LC data (0.5° × 0.5°) were resampled from 300 m resolution, which likely generated some uncertainties when calculating the global N<sub>2</sub>O budget. Furthermore, this study tends to underestimate the N<sub>2</sub>O fluxes, as a result of the inability to capture the peaks of N<sub>2</sub>O emission fluxes

**Table 5**

Sensitivity analysis of spatial estimation.

Scenarios		Total	Tropical forests	Temperate forests	Boreal forests	Grasslands
SS	COE <sub>NR</sub> value	–	0.09	0.04	0.009	0.03
	Average N <sub>2</sub> O emissions	5.02(0.17)†	3.03(0.15)	0.55(0.04)†	0.04(0.005)†	1.40(0.03)†
SPS	P + 25%	5.53(0.19)†	2.49(0.11)	1.20(0.09)†	0.05(0.008)†	1.79(0.04)†
	P – 25%	4.54(0.14)↓	3.56(0.14)	0.025(0.002)↓	0.038(0.007)↓	0.92(0.02)↓
STS	T + 0.5°C	6.33(0.22)†	3.13(0.13)	1.48(0.10)†	0.05(0.009)†	1.67(0.04)†
	T – 0.5°C	2.35(0.40)↓	1.47(0.51)↓	0.01(0.002)↓	0.004(0.0002)↓	0.86(0.06)↓
NS	COE <sub>NR</sub> value	–	0.005	0.013	0.059	0.015
	Average N <sub>2</sub> O emissions	4.29(0.15)†	2.63(0.13)	0.44(0.03)†	0.03(0.004)†	1.19(0.025)†
MS	COE <sub>NR</sub> value	–	0.014	0.044	0.124	0.044
	Average N <sub>2</sub> O emissions	5.62(0.19)†	3.77(0.16)	0.63(0.04)†	0.05(0.007)†	1.60(0.03)†

SS: standard run with mean values of COE<sub>NR</sub> for each biome region; SPS: scenario with mean values of COE<sub>NR</sub> for each biome region and the variation of air temperature; STS: scenario with mean values of COE<sub>NR</sub> for each biome region and the variation of precipitation; NS: the minimum scenario used a combination of minimum COE<sub>NR</sub> value for every biome regions; MS: maximum scenario used a combination of maximum COE<sub>NR</sub> value for every biome regions; P: daily precipitation; T: daily mean air temperature. ↓: significant decreasing trend during the period of 1992–2015; †: significant increasing trend during the period of 1992–2015; values in brackets denote the standard deviation (Tg N yr<sup>-1</sup>).

following rewetting events, especially during the period of snowmelt in spring (Zhang et al., 2017). In addition, this study considers the effects of only atmospheric CO<sub>2</sub> concentrations, climate change and land cover transition on N<sub>2</sub>O emissions and does not consider the increase in atmospheric nitrogen deposition due to human activities, which may result in the underestimation of global N<sub>2</sub>O emissions.

## 5. Conclusions

The spatial and temporal patterns of N<sub>2</sub>O fluxes from global natural forests and grasslands are estimated using the TRIPLEX-GHG model. The total N<sub>2</sub>O emission budget was calculated by considering the variations in both fluxes and land surface areas. We found that tropical regions make large contributions to the total budgets as a result of their large N<sub>2</sub>O flux values and large natural forest and grassland areas. The relatively large changes in N<sub>2</sub>O emissions among years are probably due to extreme climate events. That is, N<sub>2</sub>O emissions would increase/decrease during years with El Niño/La Niña events, which is probably related to the negative effect of precipitation on N<sub>2</sub>O fluxes in most tropical regions. However, due to the distinct wet and dry periods in tropical grasslands, El Niño/La Niña events exert different effects on N<sub>2</sub>O fluxes, which induced reductions/increases in N<sub>2</sub>O during wet periods and increases/reductions in N<sub>2</sub>O during dry periods. In addition, the results showed that soil N<sub>2</sub>O emissions from forests are positively correlated with the atmospheric N<sub>2</sub>O concentrations for only few months, and approximately 25.9% of total observed annual N<sub>2</sub>O increases in the atmosphere can be explained by the total natural emissions (forests and grasslands), which implies that although there is a rising trend in N<sub>2</sub>O emissions from natural sources (forests and grasslands in this study), other sources, including anthropogenic, riverine and oceanic N<sub>2</sub>O sources, may be the main contributors to the elevated atmospheric N<sub>2</sub>O concentrations at a short term. However, the absence of it in short term (interannual pattern) does not mean that the terrestrial biosphere does not affect the long-term trend in the atmosphere.

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## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.agrformet.2018.12.011>.

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